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An invited talk

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Published in:
The 24th International Congress on Glass - Abstracts

Publication date:
2016

Document Version
Publisher's PDF, also known as Version of record

Following the setting of glass-ionomer bio-cements and mechanical toughness, to model the cementitious process

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Bio-cements, notably glass-ionomer cements, have been in widespread use for over 40 years in dentistry and medicine. They are chemically complex, with starting ingredients comprising an alumino-fluoro-phospho-silicate glass powder and aqueous acrylic acid. The glass is also heavily phase separated: individual phases identified with alumina-silicate, Ca-F-P-rich, and Ca-F-rich phases.[1)] Progress in improving the mechanical properties of glass-ionomer cements, however, lags behind the toughness needed for permanent implants. A significant impediment to improvement has been reliance on conventional mechanical failure methods, which are necessarily retrospective.

Through the novel use of neutron Compton scattering (NCS), which directly measures atomic cohesion, we show how it is possible to follow fracture toughness *in situ* during setting. Combining NCS with differential scanning calorimetry (DSC) and coherent terahertz spectroscopy (CTS), a comprehensive picture of the setting process can be obtained. In particular fluctuations in interfacial configurations during chelation between glass and polymer are observed.[2)] Compared to conventional mechanical methods, we find that the setting of this cement is not monotonic, but characterised by abrupt features in the development of toughness, not previously directly detected (Figure 1). Moreover we find evidence that setting processes are driven by THz collective vibrations at the hybrid interface between glass and polymer (Figure 2). By harnessing Quantum Chemical calculations we are now able to show how the development and dynamics of these processes can be understood in the context of changing atomic configurations at the altering surface of the glass, all of which provides clues by which to improve the mechanical performance of bio-cements, and, by inference, other cementitious materials.

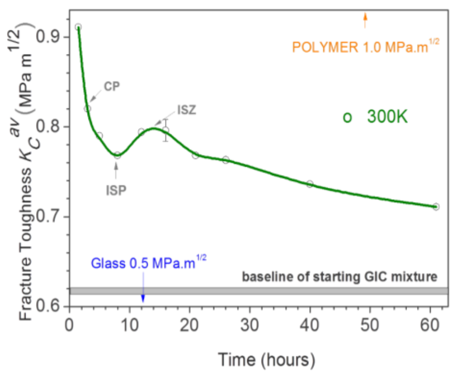

Figure 1. Developing Fracture Toughness of bio-cement as a function of setting time obtained from Neutron Compton Scattering. The Fracture Toughness of the starting Glass and Polymer are shown for comparison. CP- Coupling Point between the glass and polymer; ISP- Initial Setting Point of GIC; ISZ- Intermediate Stress Zone.

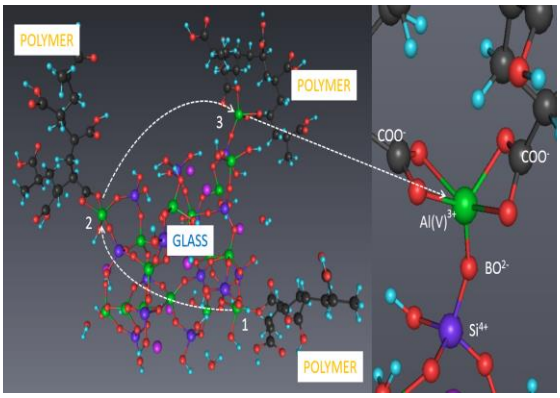

Figure 2. Geometry-optimised DFT interface model (left), identifying the glass cluster, terminated with OH^- and PAA polysalt complexes, linked via Al polyhedra. Anticlockwise sequence 1-3 shows different stages of chelation, culminating in Al(V) stable arrangement (right), modelling the hybrid interface between glass and polymer.

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